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Evidence for unmonitored coal ash spills in Sutton Lake, North Carolina: Implications for contamination of lake ecosystems

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Highlights

1. High concentrations of contaminants in coal ash pose environmental risks, particularly when coal ash leaks from storage facilities into the environment.
2. Analysis of magnetic susceptibility, visual confirmation of coal ash particles, trace element distributions, and strontium isotope ratios of bottom sediments collected in 2015 and 2018 from Sutton Lake in eastern North Carolina revealed the presence of coal ash solids mixed with natural sediments.
3. The CCR-impacted sediments exceeded ecological screening standards in fresh water lakes for multiple contaminants.
4. Porewater and leaching experiments of the CCR-impacted sediments show mobilization of soluble contaminants to the aquatic phase, potentially endanger the lake ecological system.
5. The findings from this study imply that unmonitored coal ash spills may be more common than previously realized and other lakes near coal ash storage facilities, particularly in areas susceptible to hurricane events or frequent flooding, may also be impacted.

Abstract

Coal combustion residuals (CCRs, also known as “coal ash”) contain high concentrations of toxic and carcinogenic elements that can pose ecological and human health risks upon their release to the environment. About half of the CCRs that are generated annually in the U.S. are stored in coal ash impoundments and landfills, in most cases adjacent to coal plants and waterways. Leaking of coal ash ponds and CCR spills are major environmental concerns. One factor which may impact the safety of CCRs stored in impoundments and landfills is the storage area’s predisposition to flooding. The southeastern U.S., in particular, has a large number of coal ash impoundments located in areas that are vulnerable to flooding. In order to test for the possible presence of CCR solids in lake sediments following Hurricane Florence, we analyzed the magnetic susceptibility, microscopic screening, trace element composition, and strontium isotope ratios of bottom sediments collected in 2015 and in 2018 from Sutton Lake in eastern North Carolina and compared them to a reference lake. The results suggest multiple, apparently previously unmonitored, CCR spills into Sutton Lake from adjacent CCR storage sites. The enrichment of metals in Sutton Lake sediments, particularly those with known ecological impact such as As, Se, Cu, Sb, Ni, Cd, V, and Tl, was similar to or even higher than those in stream sediments impacted by the Tennessee Valley Authority (TVA) in Kingston, Tennessee, and the Dan River, North Carolina coal ash spills, and exceeded ecological screening standards for sediments. High levels of contaminants were also found in leachates extracted from Sutton Lake sediments and co-occurring pore water, reflecting their mobilization to the ambient environment. These findings highlight the risks of large-scale unmonitored spills of coal ash solids from storage facilities following major storm events and contamination of nearby water resources throughout the southeastern U.S.

1. Introduction

The adequate storage and management of coal combustion residuals (CCRs) is a major challenge facing energy utilities in the U.S. (U.S. EPA, 2015; Punshon, 2003). CCRs represent one of the largest industrial solid waste streams in the U.S. and typically contain high concentrations of toxic and carcinogenic elements, which, upon release to the environment, could pose human health and ecological risks (Cordoba et al., 2012; Dreesen et al., 1977; Izquierdo and Querol, 2012; Kosson et al., 2002; Meij and Winkel, 2007; 2009; Nelson et al., 2010; Swaine, 1992, 1994; Thorneloe et al., 2010; Twardowska, 2003). Over 100 million tons of CCRs are generated annually; about half is reused, mostly by the cement industry (about 33%), while the other half is stored in open impoundments and landfills (U.S. EPA, 2015). Even with the reduction of coal combustion and decommissioning of coal plants in the U.S. due to the rise of shale gas (Kharecha et al., 2010), CCR storage remains a major public policy and environmental problem. There is significant evidence of leaking of CCR storage facilities and contamination of underlying groundwater and associated surface waters (Harkness et al., 2016; Rowe et al., 2002; Ruhl et al., 2012). The safe storage of CCRs in coal ash ponds and landfill sites can also be affected by natural disasters, such as hurricanes. This is particularly relevant in the southeastern U.S., where large numbers of coal ash impoundments are actively used or are in the process of being decommissioned.

Previous studies have addressed the environmental risks associated with CCR disposal through investigation of the distribution of toxic and carcinogenic elements in coals and CCRs, (Cordoba et al., 2012; Dai et al., 2014; 2018; Lauer et al., 2015; Meij and Winkel, 2009; Silva et al., 2012; Swaine, 1992; 1994; Swanson et al., 2013) by exploring the mechanisms that control

the mobilization of contaminants from CCRs, (Izquierdo and Querol, 2012; Kosson et al., 2002; Liu et al., 2013; Schwartz et al., 2016; 2016; Thorneloe et al., 2010) and via monitoring cases where associated surface water and groundwater were impacted by: (1) major coal ash spills such as the Tennessee Valley Authority (TVA) in Kingston, Tennessee (Cowan et al., 2013; 2015; Ruhl et al., 2009; 2010; 2014) and the Dan River, North Carolina (Cowan et al., 2017; Shin et al., 2017; Yang et al., 2015) spills; (2) the disposal of CCR effluents (Dreesen et al., 1977; Ruhl et al., 2012); and (3) leaking of coal ash impoundments (Harkness et al., 2016). Since the installation of high-efficiency cold-side electrostatic precipitators (ESPs), fabric filters, and wet flue gas desulphurization (FGD) in all U.S. thermoelectric plants, the conventional wisdom has been that any CCR environmental impact is related to either long-term fluid leaking from inadequate CCR storage infrastructure or acute impacts from major infrastructure failure and spills (Harkness et al., 2016; Lemly, 2018; Lemly and Skorupa, 2012; Ruhl et al., 2009; 2010; 2012). This study presents evidence for the presence of CCR solids in lake bottom sediments and contamination of the aquatic system. CCR transport from coal ash ponds to the adjacent lakes could result from flooding, such as happened during Hurricane Florence in 2018, although other mechanisms such as unintentional CCR release and past dumping practices or historic CCR placement in the lake cannot be ruled out.

This study presents the risks for unmonitored CCR spills through investigation of Sutton Lake near Wilmington, North Carolina (Fig. 1) and demonstrates the vulnerability of decommissioned CCR storage sites to hurricane events and the potential of CCR transport to nearby water resources. Since the early 1970's, a 4.45 km² impoundment known as Sutton Lake was used for cooling the nearby Sutton coal-fired steam plant. In 2013, the coal-fired plant was retired and replaced with a 625-MW natural gas combined-cycle plant. The CCRs, which have

been generated for decades, have been stored in impoundments and a landfill adjacent to Sutton Lake, which is widely used for boating and fishing, and plays host to abundant wildlife. In September 2018, a Category 4 major hurricane (Hurricane Florence) hit eastern North Carolina and caused significant flooding of the major river systems, including the Cape Fear River adjacent to Sutton Lake. The flooding caused a breach of the barrier between Sutton Lake and the Cape Fear River, allowing flow of the upstream Cape Fear River through Sutton Lake and back to the downstream river (Fig. 1). The possible transport of CCRs from the nearby impoundment and landfill became a major public concern, yet those concerns were not confirmed with independent testing for the presence of CCRs in the lake and river sediments.

In order to determine the possible presence of CCR particles in Sutton Lake, multiple geochemical and physical diagnostic proxies were used to analyze the lake-bottom sediments. Previous studies have shown that CCRs are enriched in several trace metals (e.g., boron, arsenic, selenium, molybdenum) (Fletcher et al., 2014; Harkness et al., 2016; Izquierdo and Querol, 2012; Ruhl et al., 2009; 2010; 2012; Schwartz et al., 2016; 2018; Swanson et al., 2013; Tian et al., 2018; Twardowska, 2003; Zhao et al., 2018), have a distinctive strontium isotope fingerprint (Ruhl et al., 2014), and are characterized by elevated magnetic susceptibility resulting from combustion of pyrite-containing coals (Cowan et al., 2013; 2015; 2017; Flanders, 1999; Grimley et al., 2017; Gune et al., 2016). In this study, these different and independent proxies were integrated to evaluate the possible presence of CCRs in the bottom sediments of Sutton Lake compared to sediments from an upstream portion of the Cape Fear River and a reference lake (Lake Waccamaw) that is not associated with CCR disposal. In addition, the mobilization of various elements from CCR-impacted sediments were evaluated through differential leaching experiments and via measurement of trace elements in pore water associated with impacted

sediments. Given that contamination of sediment pore water can trigger bioaccumulation through the ecosystem (Brandt et al., 2017; 2018), this could have grave implications for ecosystem health.

2. Methodology

2.1. Sediments sources and sampling sites

This study focuses on Sutton Lake, which for decades (1972-2013) was used as cooling water for and received CCR effluents from the nearby L.V. Sutton Steam Plant. In November 2013, the coal-fired units were retired and replaced by with a 625-MW natural gas combined-cycle plant. We also studied Lake Waccamaw in coastal North Carolina (Fig. 1) which was used as a reference lake without known CCR impact. On October 22nd 2018, we collected sediments samples within near-surface sediments from the lake at seven sites in Sutton Lake and three in the Cape Fear River (Fig. 1). We also studied three sediment samples from Sutton Lake and three sediment samples from Lake Waccamaw that were collected in 2015 (Brandt, 2018; Brandt et al., 2017; 2018). The 2015 samples were collected as part of the previous study and archived, and then analyzed along with the 2018 samples collected from this study. Lake sediments were collected using a box corer, transported back to Duke University, and dried and homogenized within 48 hours.

2.2. Analytical procedures

Sediment samples were processed via (1) full digestion using HF-nitric combined acids for total dissolution of the sediments; (2) leaching with DI water in a 1:10 ratio for water extraction of soluble elements; (3) leaching in 1N HNO₃; (4) strontium isotopes measurements; and (5) frequency-dependent mass-specific magnetic susceptibility measurement.

2.2.1. Magnetic susceptibility

Frequency-dependent mass-specific magnetic susceptibility (χ) was measured at low (K_{LF} 0.46 KHz) and high (K_{HF} 4.6 KHz) frequencies on 6.02-cm³ plastic cubes packed with dry sediment using a Bartington Instruments MS-3 meter with a dual-frequency MS2B sensor at Appalachian State University (Cowan et al., 2013; 2015; 2017). The volume-specific magnetic susceptibility values (K) were converted into mass specific susceptibility (χ) to account for

samples with different densities. The volume-magnetic susceptibility (K) is divided by the bulk density of the sample to obtain a mass-specific magnetic susceptibility expressed in units of m^3/kg (Dearing, 1999). Percent frequency-dependent magnetic susceptibility ($\chi_{\text{FD}}\%$) is calculated as: $\chi_{\text{FD}}\% = (\chi_{\text{LF}} - \chi_{\text{HF}} / \chi_{\text{LF}}) \times 100$. A high $\chi_{\text{FD}}\%$ indicates the presence of ultrafine ($<0.03\ \mu\text{m}$) superparamagnetic ferromagnetic minerals (Dearing, 1999), typical for soils, whereas fly ash typically has low $\chi_{\text{FD}}\%$ (Magiera et al., 2011). The percent ash in each sample measured for χ was determined by point counting using a Leica DMLP polarizing microscope with a Swift model F automated point counter. Smear slides were made by taking a small amount of dry sample from the χ cube and distributing it in distilled water across a 27-mm x 49-mm glass slide. Upon drying, the sample was permanently mounted under a coverslip with epoxy having a refractive index of 1.520 (Loctite Impruv 363). Slides were counted under 200x magnification using a standard point counting method devised to quantify ash within riverbed sediment at the TVA Kingston spill (RJ Lee Group, 2010). Only particles that fell under the crosshairs were counted to reach 300 counts. Particles were identified as coal ash based on Fisher et al. (1978) and included spheres, amorphous ash, and lacy particles. Mineral grains included clay-size particles as well as silt and sand grains. Organic matter included fibrous plant fragments as well as freshwater microfossils, mostly diatoms. If the crosshairs landed on an empty space or if the particle could not be assigned to one of the above groups, the stage was advanced to the next point.

2.2.2. *Sediments extraction*

34 ± 1 mg of sediment samples were weighed in 10-mL Teflon vials and digested overnight at 90–100 °C on a hotplate in a HF-HNO₃ mixture (v/v=3 mL: 2 mL; optimal grade). The digested samples were then dried down completely and re-digested overnight at 90–100 °C in a mixture of 15-M HNO₃ (1 mL), H₂O₂ (1 mL; Optima grade), and quartz-distilled (QD) water (5 mL). Following the re-digestion, 0.2-mL aliquot of each digest (7 mL in total) was diluted to 2 mL for the measurement of trace element concentrations on a VG PlasmaQuad-3 inductively coupled plasma mass spectrometry (ICP-MS). The accuracy was assessed by measuring the National Institute of Standards and Technology (NIST) standard reference material (SRM) for trace elements in coal fly ash, SRM 1633a (Table 1).

2.2.3. *Leaching procedure*

Each sample underwent two leaching procedures, one using DI water and one using 1-M Optima Nitric Acid. Samples were leached in 50-mL centrifuge tubes, with about 4 grams of solid and 40 grams of liquid being used in each experiment. Tubes were placed on a New Brunswick Scientific C1 Platform Shaker and mixed at 180 rpm for 24 hours. Each leachate was extracted using a 30-mL syringe, filtered using 0.45- μ m syringe filters, and collected in a 60-mL acid washed HDPE bottle. DI leachates were then acidified using 7 molar Optima Nitric Acid to pH 2 before analysis by ICP-MS. To remove organic interference, 1 mL of digestate was added to 800 μ L of 15N-nitric acid in Teflon vials. The mixture was then capped and heated at 100 C on a hotplate for 48 hours after which 200 μ L of hydrogen peroxide was added to the mixture and allowed to react.

2.2.4. Analytical procedure

Trace elements were analyzed by inductively coupled plasma mass spectrometry (ICP-MS) on a VG PlasmaQuad-3 after gravimetric dilution in 2% HNO₃. Detection limits were evaluated by multiplying the standard deviation of repeated blank measurements by three and dividing by the slope of the external standard. Strontium was separated using Eichrom Sr-specific ion exchange resin. ⁸⁷Sr/⁸⁶Sr ratios were measured on a Thermo Fisher Triton thermal ionization mass spectrometer at Duke University in positive mode using single Re filaments with a precision of ± 0.000006 on repeated measurements of NIST SRM 987 standard (mean=0.71062, n=98).

3. Results

3.1. Evidence for coal ash solids in bottom sediments of Sutton Lake

The presence of CCRs in Sutton Lake sediments (Fig. 1) was evaluated by testing the magnetic susceptibility and geochemistry of the sediments. Previous studies have shown that combustion of coal containing iron sulfide (pyrite and marcasite) causes the formation of magnetic minerals such as spherical magnetite in CCRs (Flanders, 1999). Low-field magnetic susceptibility (χ_{LF}) was shown to be strongly positively correlated with total coal ash in riverbed samples impacted by previous CCR spills at the Kingston Fossil Plant (TVA) (Cowan et al.,

2015) and Dan River Steam Plant (Duke Energy) (Cowan et al., 2017), with low frequency dependence commonly reflecting presence of coal ash (Magiera et al., 2011). The magnetic susceptibility measured in sediments from Sutton Lake ranged over three orders of magnitude, from $6.06 \times 10^{-9} \text{ m}^3/\text{kg}$ for sediments containing no visual evidence of coal ash to $6.21 \times 10^{-6} \text{ m}^3/\text{kg}$ for sediments with visual evidence of coal ash (Table 2). Coal ash particles were optically observed in sediment samples and each contained a mix of spherical ash and non-spherical amorphous particles (Fig 2). All observed morphologies in sediments from Sutton Lake were previously described as components of fly ash (Fisher et al., 1978; Hower, 2012) and were found in our previous studies (Cowan et al., 2013; 2017). High correlation ($R^2 = 0.95$; $p < 0.001$) between χ_{LF} and observed percentage of fly ash particles (Fig. 2) reflects the presence and mixing of CCRs with naturally occurring sediments in Sutton Lake. The effectiveness of detecting CCRs with distinctive χ_{LF} at Sutton Lake sediments is aided by the high abundances of quartz and CaCO_3 minerals as well as organic matter in the sediment, which do not contribute to the magnetic signal of the bulk sediments.

A second line of independent evidence is the distribution of trace elements in Sutton Lake sediments. The concentrations of trace elements in Sutton Lake sediments and a coal fly ash standard (NIST 1633c) were compared to those in sediments from Lake Waccamaw, which is used as a reference (non-impacted) lake since it is located in similar geological setting without coal ash input (Table 3). Sediments from Sutton Lake are enriched by one to two orders of magnitude in CCR-related elements relative to those in Lake Waccamaw, with conspicuously high enrichments of Cu, As, Se, Mo, Sb, and Tl (Figs. 2 and 3). Those elements are known to be enriched in CCRs (Cordoba et al., 2012; Dreesen et al., 1977; Fletcher et al., 2014; Harkness et al., 2016; Izquierdo and Querol, 2012; Kosson et al., 2002; Meij and Winkel, 2007; 2009; Rowe

et al., 2002; Schwartz et al., 2016; 2018; Swaine, 1992; 1994; Swanson et al., 2013; Thorneloe et al., 2010; Tian et al., 2018; Twardowska, 2003; Zhao et al., 2018), which is consistent with the enrichment measured in the fly ash standard, providing further support for the presence and mixing of CCR solids in Sutton Lake sediments. The observed high correlations between the concentrations of these elements (i.e., Se versus As $R^2=0.82$, $p=0.008$; $n=6$; Sb vs. Mo $R^2=0.53$, $p=0.06$; $n=6$; Fig. 2), indicate mixing between CCRs and naturally occurring sediments.

A third line of evidence for the presence of CCRs in Lake Sutton sediments is the distinctive Sr isotope ratio of the Sutton Lake sediments. The $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of sediments from Lake Sutton (full digestion) varied between 0.71129 to 0.71386, which overlaps with the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios reported for the Appalachian coals (0.7117 to 0.7126; Fig. 4) (Vengosh et al., 2013), which have been used for North Carolina coal plants (Ruhl et al., 2012).

Thus, combined, there are three independent indicators for the presence of CCRs in Sutton Lake sediments after Hurricane Florence. During Hurricane Florence, the flow rates in the Cape Fear River exceeded the annual flow rates by 10-fold (Fig. 5), which resulted in the river overflowing through the lake and surrounding areas, including the adjacent coal ash pond and landfills (Fig. 1). The data from our sampling at different sites in the lake suggest transport and deposition of CCRs particles in different parts of the lake. Downstream from Sutton Lake, sediments in Cape Fear River (Fig. 1; $n=2$) show relatively low metals concentrations compared to the lake sediments. The downstream Cape Fear River sediments had slightly higher concentrations of Ni, Cu, and As relative to those in sediments from the upstream Cape Fear River (Table 3), which could reflect very small presence of CCRs solids in the downstream river sediments. Further investigation is needed to fully evaluate the possible migration of CCRs solids downstream in the Cape Fear River.

The sample analysis in this study also included the lake sediments from Sutton Lake collected by Brandt et al. (2015) and Brandt (2018) two years after the decommissioning of the Sutton Coal Plant (Table 3). Similar to the lines of evidence for the presence of CCRs in the lake sediments post Hurricane Florence, the data show that the 2015 Sutton Lake sediments also contained CCR solids. This is demonstrated by the elevated magnetic susceptibility (Fig. 2A) and trace metals distribution that mimic the abundance of metals in fly ash standard as normalized to the reference lake (Figs. 2 and 3). A comparison of the mean values of trace elements measured in Sutton Lake in 2018 relative to the mean values in sediments from Sutton Lake collected in 2015 shows similar concentrations (Table 4). These results suggest that CCR solids were previously (i.e., prior to 2018 flooding event) transported to the lake and accumulated in different parts of Sutton Lake (Fig. 1), regardless of the operation of the coal plant, which was already phasing out in 2013. Figure 5 presents the flow rates in the Cape Fear River during the last 35 years (USGS dataset, 2018) and demonstrates that Hurricane Florence was not alone in generating high river flows above baseline, as many other storm events have also generated abnormal high flow rates that could result in the flooding of adjacent CCR storage facilities and the transport of CCR particles into Sutton Lake.

While we link the presence of CCRs in bottom sediments of Sutton Lake to flooding events, CCR transport as a result of human errors cannot be excluded; the close proximity of the landfill and coal ash ponds to the lake (Fig. 1), coupled with operation activities, both routine and related to the transition from impoundments to landfills, could also result in unreported CCRs spills. Likewise, historic CCR disposal practices at the site cannot be excluded. In order to test the likelihood of CCRs mobilization as part of the routine operations, data of annual fluxes of effluents discharge from the outfall of the coal ash pond in Sutton Lake and the concentrations

of solids reported in these effluents were examined. Mass-balance calculations for possible transport of CCR solids from the outfall of Sutton Lake using data of North Carolina Department of Environment and Natural Resources (2018) (Table 5) indicate that effluents containing total suspended solids (TSS) of 4.5 mg/L (the mean value during 2010-2013; North Carolina Department of Environment and Natural Resources, 2018) with known discharge rates (mean value of 6.4×10^9 L per year) could not have any meaningful contribution to the mass of the upper 25 cm of Sutton Lake bottom sediments (Table 5). Therefore, it seems that CCR transport to Sutton Lake was not from continuous discharge of CCR solids in effluents under routine operation conditions.

3.2. *Contaminants in sediments and mobilization to the ecosystem*

Metal concentrations in Sutton Lake sediments collected in 2015 and 2018 were enriched by one to two orders of magnitude compared to their concentrations in the reference lake (Lake Waccamaw) (Figs. 3 and 6). The concentrations of contaminants known to have ecological impacts found in Sutton Lake sediments were lower (As), similar (Cd), or even higher (Se, Ni, Sb, Cu, V, Co, Pb) than those measured in impacted river sediments from the Kingston, Tennessee (Tetra Tech Report, 2008) and the Dan River, North Carolina (N.C. Water Quality Report, 2018) coal ash spills (Fig. 6), implying a serious ecological threat, comparable to previous known coal ash spills. Metal concentrations found in Sutton Lake sediments exceeded the freshwater sediment screening benchmarks values (Fig. 6) developed to define fresh-water-lake toxicity potential (Ingersoll et al., 2001; Long et al., 2006; Long et al., 2000; MacDonald et al., 2000; MacDonald, 2003), which are commonly used by the U.S. EPA as sediment quality guidelines in evaluating threshold-effect concentrations (MacDonald, 2003).

Previous studies have shown that the conditions prevailing on the lake or river sediments would control the mobilization of contaminants from solid CCRs into the ambient aquatic system. In particular, the redox state would control redox-sensitive elements. For example, under reducing conditions elements like As would preferentially mobilize and become enriched in pore water associated with CCRs. In contrast, under oxidizing conditions, elements like Se are preferentially mobilized (Ruhl et al., 2010; Schwartz et al., 2016; 2018). Other factors such as the pH would determine the leachability of metals (low-pH) relative to metalloids (high-pH). Thus, combined oxidation-reduction potential (ORP) and pH would determine the speciation and mobility of elements from CCRs (Schwartz et al., 2016; 2018).

In addition to the measurement of metals in the sediments (total digestion), leaching experiments were conducted in order to test the relationships between the occurrence of metals in the bulk sediments to metals in water and acid (1 N nitric acid) leachates. The results show that the concentrations of trace elements in the water and acid leachates generally follow the concentrations of the bulk sediments (Table 6). A comparison of the concentrations of trace elements in the leachates from Sutton Lake sediments to those in leachates of sediments from the reference lake (Lake Waccamaw), shows one- to two-orders of magnitude enrichments of trace metals in both water and acid leachates in Sutton Lake sediments relative to those in Lake Waccamaw. Similar enrichment factors were observed for metal concentrations in bulk sediments from the two lakes. Furthermore, the enrichment factor distribution pattern in the water leachates (Fig. 3) mimics the patterns observed for the bulk sediments, reflecting the high reactivity of the Sutton Lake sediments and the mobilization of toxic elements that are known to be enriched in CCRs.

In addition, pore water data collected in 2015 from Sutton Lake and Lake Waccamaw reported in Brandt et al. (2018) and Brandt (2018) used to further evaluate the mobilization of CCR-contaminants to the ambient environment. The data show that trace metal concentrations in the pore water extracted from the 2015 Lake Sutton sediments (Brandt, 2018) were systematically higher than those in pore water collected from Lake Waccamaw sediments (Fig. 3). Several of the metals that were conspicuously enriched in the bulk sediments and water and acid leachates were also highly enriched in the pore water, including Cu, Mo As, Se, Sb, and Tl (Fig. 3). These results are consistent with high levels of CCR-contaminants measured in pore water from sediments impacted by the 2008 Kingston TVA coal ash spill into the Emory River, Tennessee (Ruhl et al., 2010). The pore water extracted from Emory River sediments covered by coal ash downstream from the spill site had elevated arsenic (up to 2000 µg/L) and boron (Ruhl et al., 2010). While the concentrations of B and As in the pore water from Sutton Lake (175 and 18.7 µg/L, respectively) collected in 2015 (Brandt, 2018) were lower than those measured in the Kingston spill impacted river, they were significantly higher than pore water from the background lake (17.5 and 1.1 µg/L, respectively) reflecting the mobilization of CCR-contaminants and the potential of negative impact of the spilled CCRs solids on the ambient environment.

The Sr isotope ratios obtained from the bulk Sutton Lake sediments, water leachates from Sutton Lake sediments, 2015 Sutton pore water, 2015 Lake Waccamaw sediment water leachates, and 2015 Lake Waccamaw pore water were compared (Fig. 4). The $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of the water leachates of Sutton Lake (0.710082 ± 0.0002) were lower than those of the bulk sediments (0.71257 ± 0.00107 ; Fig. 4), reflecting selective mobilization towards a lower $^{87}\text{Sr}/^{86}\text{Sr}$ ratio in the water-soluble Sr. The Sr isotope ratio of the water-soluble Sr was identical to the

$^{87}\text{Sr}/^{86}\text{Sr}$ ratios measured in Sutton Lake in 2015 ($0.71008 \pm 8 \times 10^{-5}$) (Brandt et al., 2018), suggesting a similar differential Sr isotope leachability. Likewise, the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio in water-soluble Sr from Waccamaw sediments is identical to the ratio in pore water collected from Lake Waccamaw in 2015 (Fig. 4). Consequently, the Sr isotope data indicate that the occurrence of CCRs in Sutton Lake sediment resulted in mobilization of Sr with a higher $^{87}\text{Sr}/^{86}\text{Sr}$ ratio to the aquatic phase relative to the Sr isotope ratio in the pore water from the reference lake. The observation of differential modification of lower $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in the aquatic phase relative to the bulk sediments is consistent with the results of leaching experiments conducted in CCRs originated from Appalachian coals (Ruhl et al., 2014).

Data from the leaching experiments conducted in this study show high mobilization of many of the trace elements known to be enriched in CCRs (Fig. 3), which is consistent with the known distribution of trace elements in CCRs (Meij and Winkel, 2007; 2009; Swaine, 1992; 1994; Swanson et al., 2013; Thorneloe et al., 2010; Tian et al., 2018; Twardowska, 2003). The mobilization and enrichment of toxic elements in co-existing pore water could induce bioaccumulation of toxic metals (U.S. EPA, 2015; Fletcher et al., 2014; Greeley et al., 2016; Rowe et al., 2002) as demonstrated by high Se concentrations in fish tissues (Brandt et al., 2017) and Sr isotope ratios in fish otoliths (Brandt et al., 2018). Consequently, chronic migration of CCR solids to lakes, as demonstrated from Sutton Lake, would result in long-term bioaccumulation in the ecological system.

4. Discussion

Ruhl et al. (2012) showed that pore water from lakes in North Carolina located near coal ash ponds and impacted by discharge of CCR effluents had systematically elevated CCR-

contaminant levels such as B and As compared to pore water from a reference lake without CCR input. They suggested that the elevated levels of CCR-contaminants in the pore water (Fig. 7; see location in Fig. 1) originated from discharge of the CCR effluents from the nearby coal ash impoundments (Ruhl et al., 2012). Likewise, several other studies have highlighted the negative impact of the discharge of effluents from coal ash ponds outfalls and the bioaccumulation of toxic elements like Se due to the discharge of CCR-enriched effluents (Brandt, 2018; Brandt et al., 2017; Lemly, 2018; Lemly and Skorupa, 2012). Yet the data presented in this study suggest that transport and accumulation of CCR solids in the sediments of impacted lakes, followed by mobilization of CCR-contaminants to the pore water could also cause this contamination. Based on the presence of CCR solids in Sutton Lake in 2015, two years after the decommissioning of the coal plant, combined with large-scale occurrence of CCR-contaminants in pore water across North Carolina (Ruhl et al., 2012) shown in Fig. 5, it is hypothesized that the case of Sutton Lake may be not unique and that CCR solids may have been transported from disposal sites and accumulated in adjacent lakes at many additional sites throughout the southeastern U.S.

Figure 1 presents the paths of major hurricanes during the last two decades, demonstrating the vulnerability of CCR disposal sites to major flooding events like Hurricane Florence in 2018. Likewise, hydrographs from major river systems in North Carolina show systematically high flow rates in major river systems of North Carolina during the last 30 years (Fig. 5), reflecting multiple weather events that could cause flooding and CCR solids transport to adjacent lakes. Observations in Sutton Lake and pore water data (Ruhl et al., 2012) from lake sediments all over North Carolina (Fig. 5) indicate a much wider scale phenomenon; CCRs are not restricted to designated disposal sites such as impoundments and landfills but are also present in lakes

adjacent to these disposal sites. This implies that the distribution and impact of CCRs in the environment is far larger than previously thought. The high concentrations of toxic metals above the acceptable ecological thresholds we found in Sutton Lake sediments requires protection and remediation measures, especially due to the extensive use of Sutton Lake for fishing and recreation. Future studies should look to test the hypothesis that other lake systems in the southeast near CCR disposal facilities contain significant CCR solids and evaluation of the potential ecological and human health implications.

5. Conclusions

This study presents new data that show evidence for the presence of coal ash solids in sediments from Sutton Lake in eastern North Carolina. The variations of magnetic susceptibility, trace metals distribution, and strontium isotope ratios suggest mixing of CCRs solids and the local sediments at different locations in the lake. We found evidence for the presence of CCR solids in near-surface sediments from the lake in samples collected in 2015 and 2018 (post Hurricane Florence), and suggest that flooding events may have caused the transport of CCRs solids from the adjacent CCR storage sites near the lake and accumulation in the lake-bottom sediments. Other mechanisms such as unintentional CCR release and past dumping practices or historic CCR placement in the lake cannot be ruled out. The high concentrations of several contaminants in Sutton Lake sediments are similar to, and even exceed, for some elements, the concentrations of contaminants in impacted sediments reported for previous coal ash spills like the Kingston and the Dan River spills, and were also above the regulated ecological guidelines for contaminants in sediments from freshwater lakes. The unmonitored spills of CCRs solids further resulted in mobilization of soluble contaminants to the aquatic phase and enrichment in

the pore water as compared to pore water extracted from a reference lake without a CCR impact. The association of CCR solids in lake sediments and pore water contamination demonstrated in this study implies that unmonitored spills may have occurred also in other lakes near CCR storage facilities, where pore water was previously shown to be elevated in CCRs associated contaminants. The possible widespread transport of CCRs solids to the environment beyond storage facilities is of concern, and future studies should verify whether water resources throughout the southeastern U.S and elsewhere are impacted by unmonitored coal ash spills.

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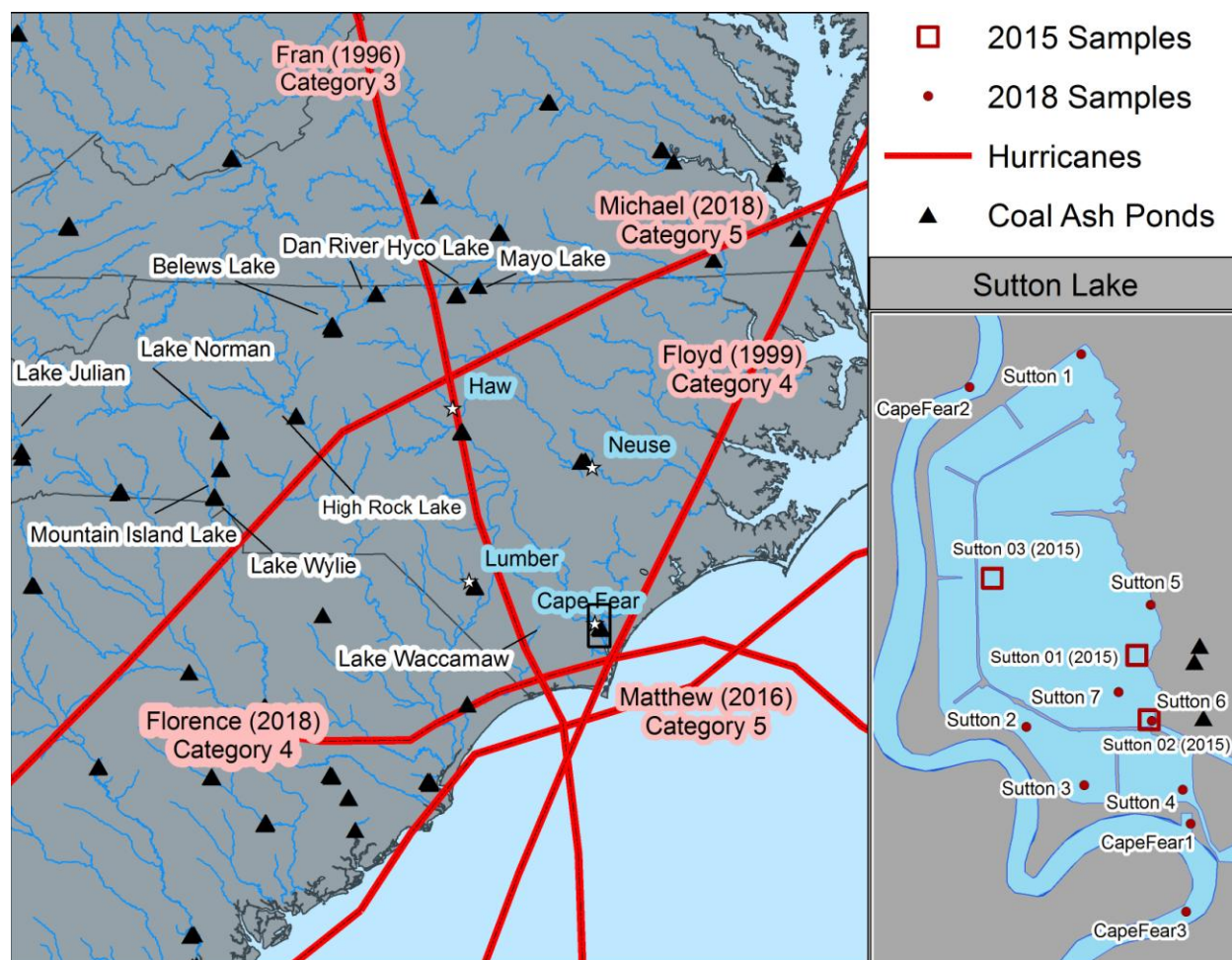


Figure 1: Map of coal ash ponds in southeastern U.S. and major hurricane tracks (> category 3) during the last two decades. Insert map shows the sampling sites locations in Sutton Lake collected in 2015 and 2018. The location of coal ash storage facilities in North Carolina is also included. Data on hurricane flow path were from the National Oceanic and Atmospheric Administration (2018). Stars on the map show locations where hydrographs were created for in figure 5.

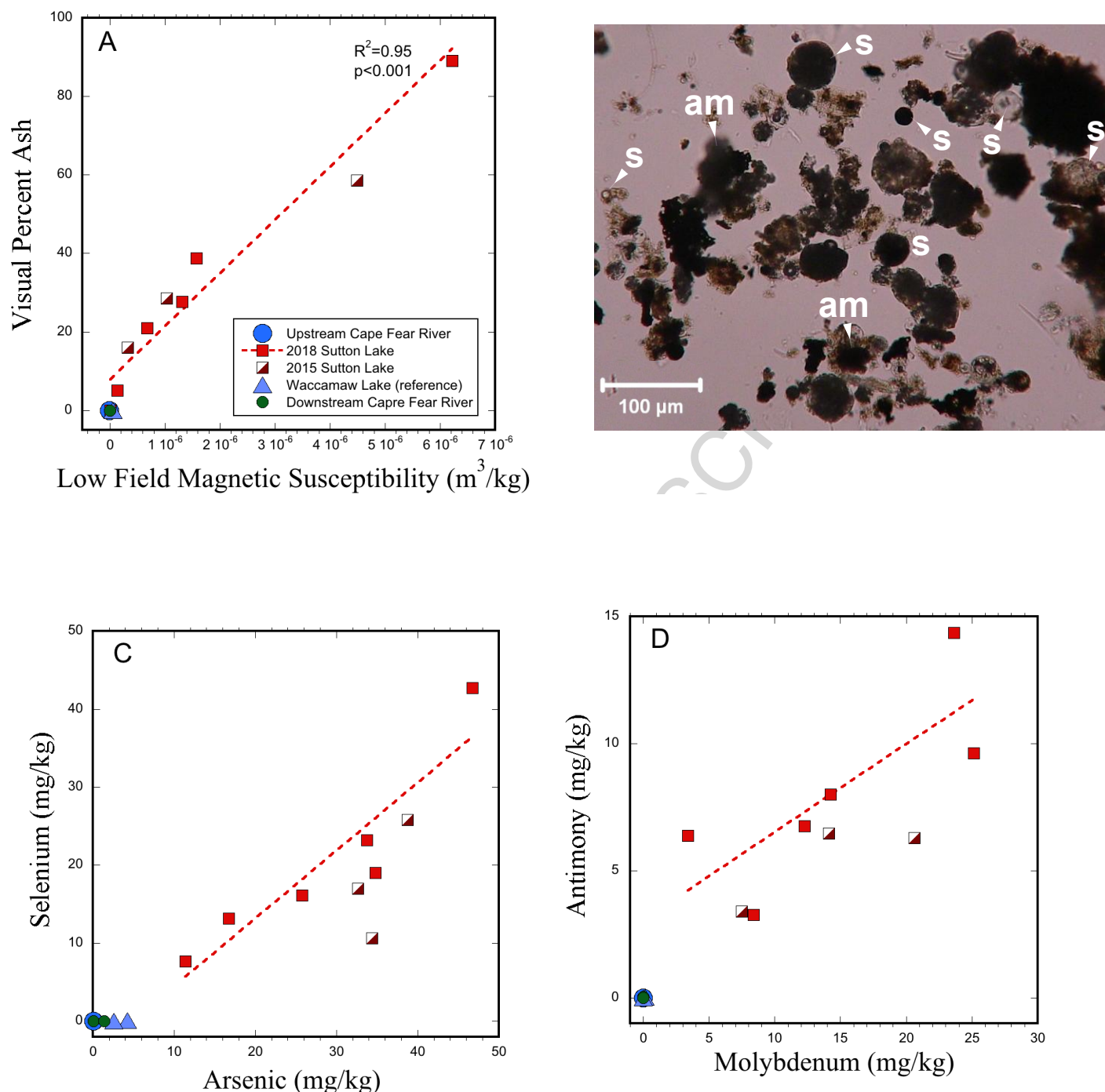


Figure 2: Multiple lines of evidence for the occurrence of CCR solids in Sutton Lake sediments: A: Percent of identified fly ash versus measured magnetic susceptibility in sediments collected from different sources.; B: Light micrograph of Sutton 03 sample with examples of ash spheres (s) and amorphous opaque (am) nonspherical ash; C: Selenium versus arsenic concentrations in sediments from different sources; D: Antimony versus molybdenum concentrations in sediments from different sources.

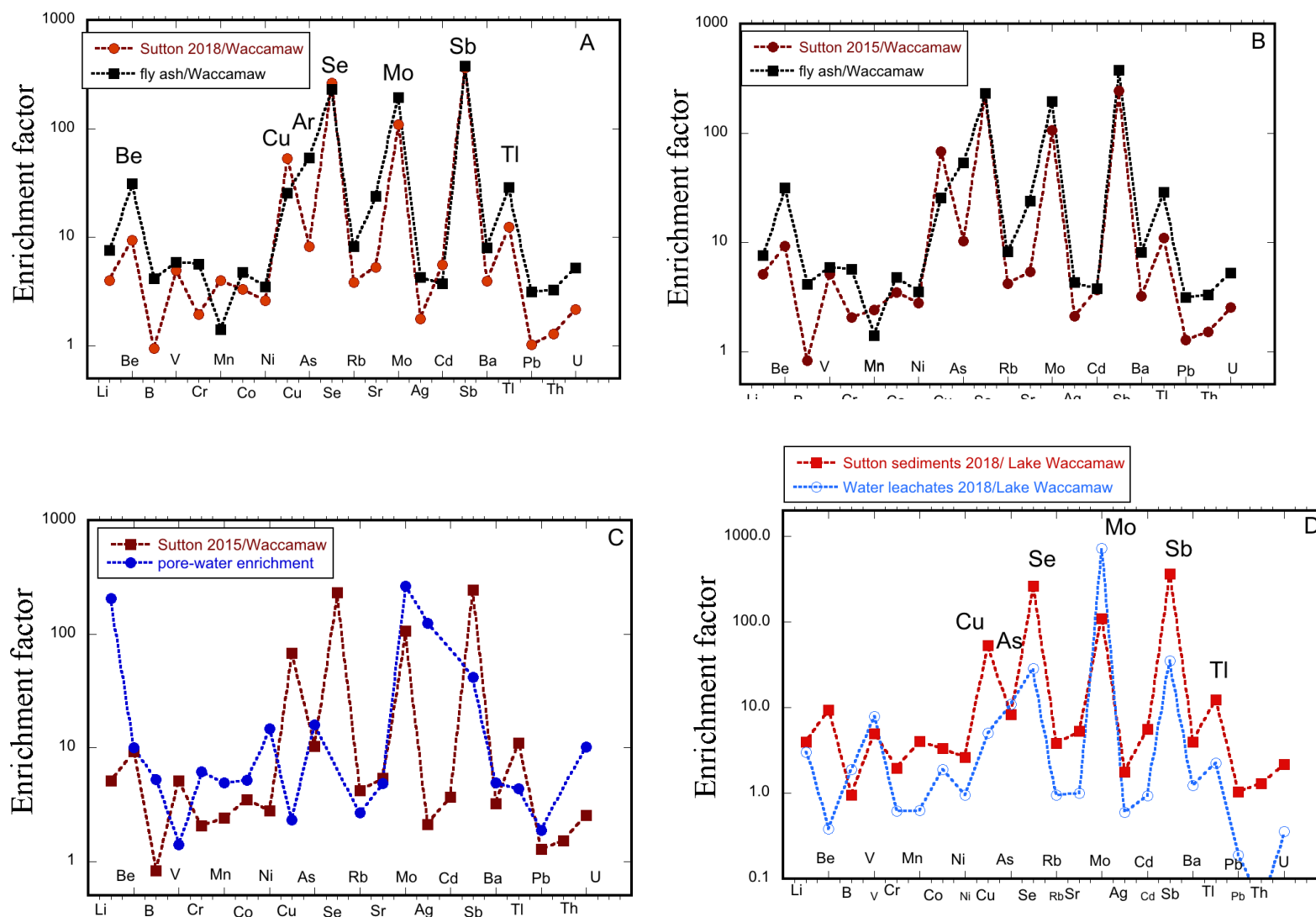


Figure 3: Distribution and enrichment of trace elements in sediments (A and B), pore water (C), and leachates (D) relative to the trace element concentrations of sediments, pore water, and leachates in the reference lake (Lake Waccamaw). A: Mean values of metals in Sutton lake sediments collected in 2018 and fly ash standard (NIST SRM 1633c, labeled “CCR”) compared to metals concentrations in Lake Waccamaw; B: Mean values of metals in Sutton lake sediments collected in 2015 and fly ash standard (NIST SRM 1633c, labeled “CCR”) compared to metals concentrations in Lake Waccamaw; C: Mean values of metals in pore water and Sutton lake sediments collected in 2015 compared to metals concentrations in Lake Waccamaw sediments and pore water (data from Brandt, 2018); and D: Mean values of metals in water leachates extracted from Sutton Lake sediments and 2018 Sutton Lake sediments compared to water leachates and bulk sediments from Lake Waccamaw.

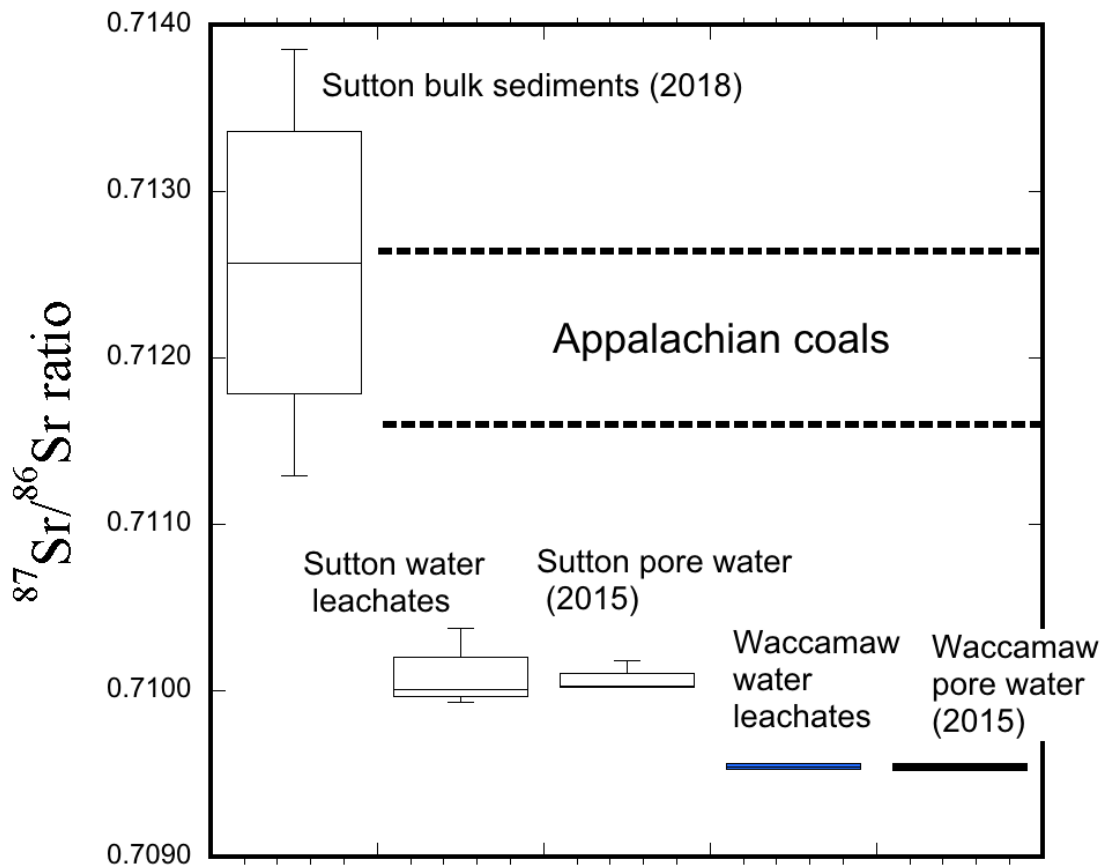


Figure 4: Box plot of Sr isotope variations of Sutton Lake sediments, water leachates extracted from Sutton Lake sediments, Sutton pore water collected in 2015, Waccamaw Lake sediments, and Waccamaw pore water collected in 2015. The Sr isotope data indicate that (1) sediments from Sutton Lake have a $^{87}\text{Sr}/^{86}\text{Sr}$ ratio that mimic the ratios reported for the Appalachian coals (Vengosh et al., 2013); and (2) that Sr isotope ratios in water leachates are identical to pore water in both the Sutton and Waccamaw lakes; and (3) selective mobilization of Sr to the water phase with a lower $^{87}\text{Sr}/^{86}\text{Sr}$ ratio relative to the $^{87}\text{Sr}/^{86}\text{Sr}$ in the bulk sediments. Sr isotope ratios in Sutton Lake and Lake Waccamaw pore waters from 2015 are from Brandt et al. (2018).

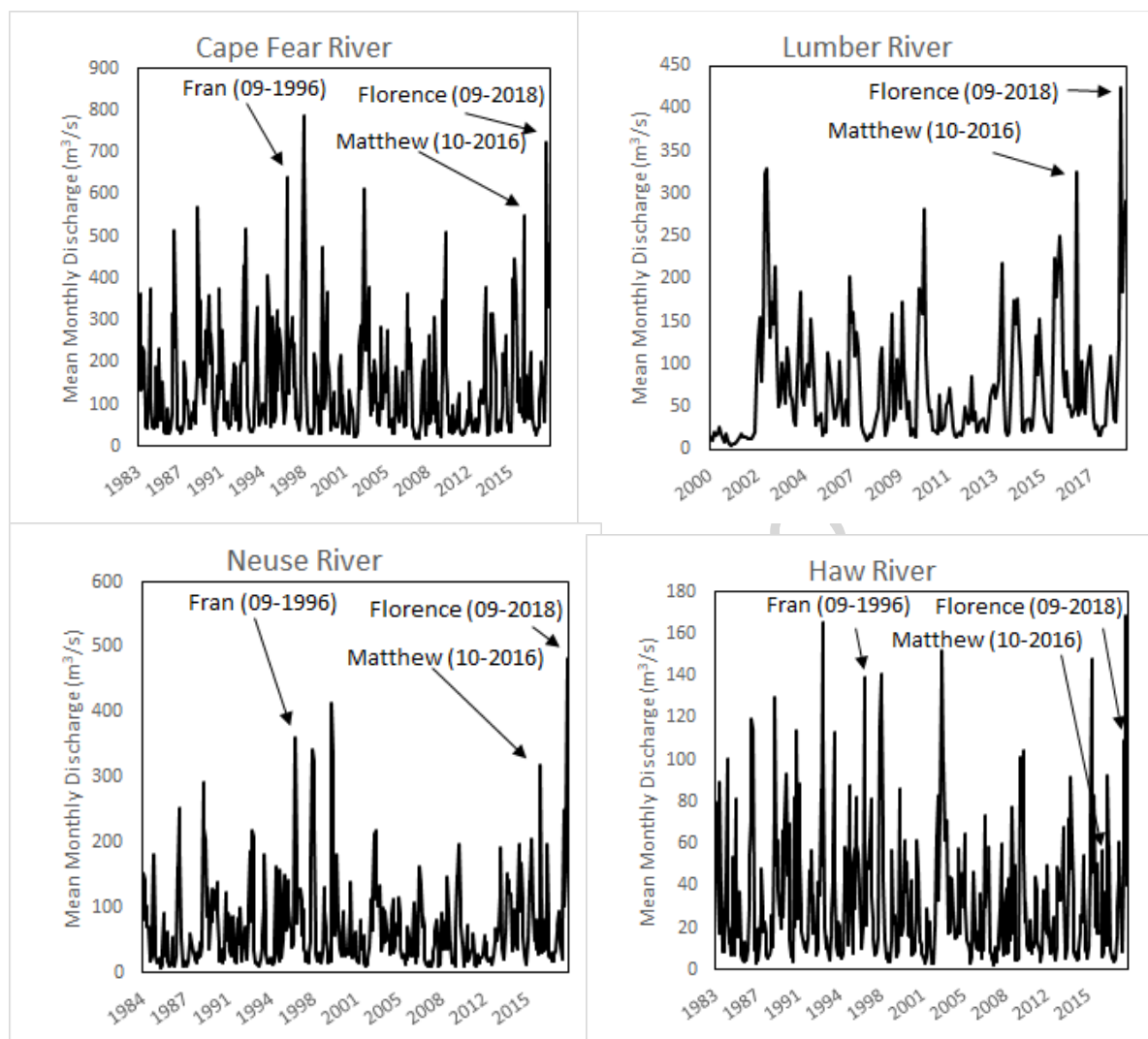


Figure 5: Hydrographs of mean monthly discharge rates (m^3/s) of major river systems in North Carolina (see locations in Fig. 1). Data was generated from U.S. Geological Survey Current Water Data for North Carolina (2018).

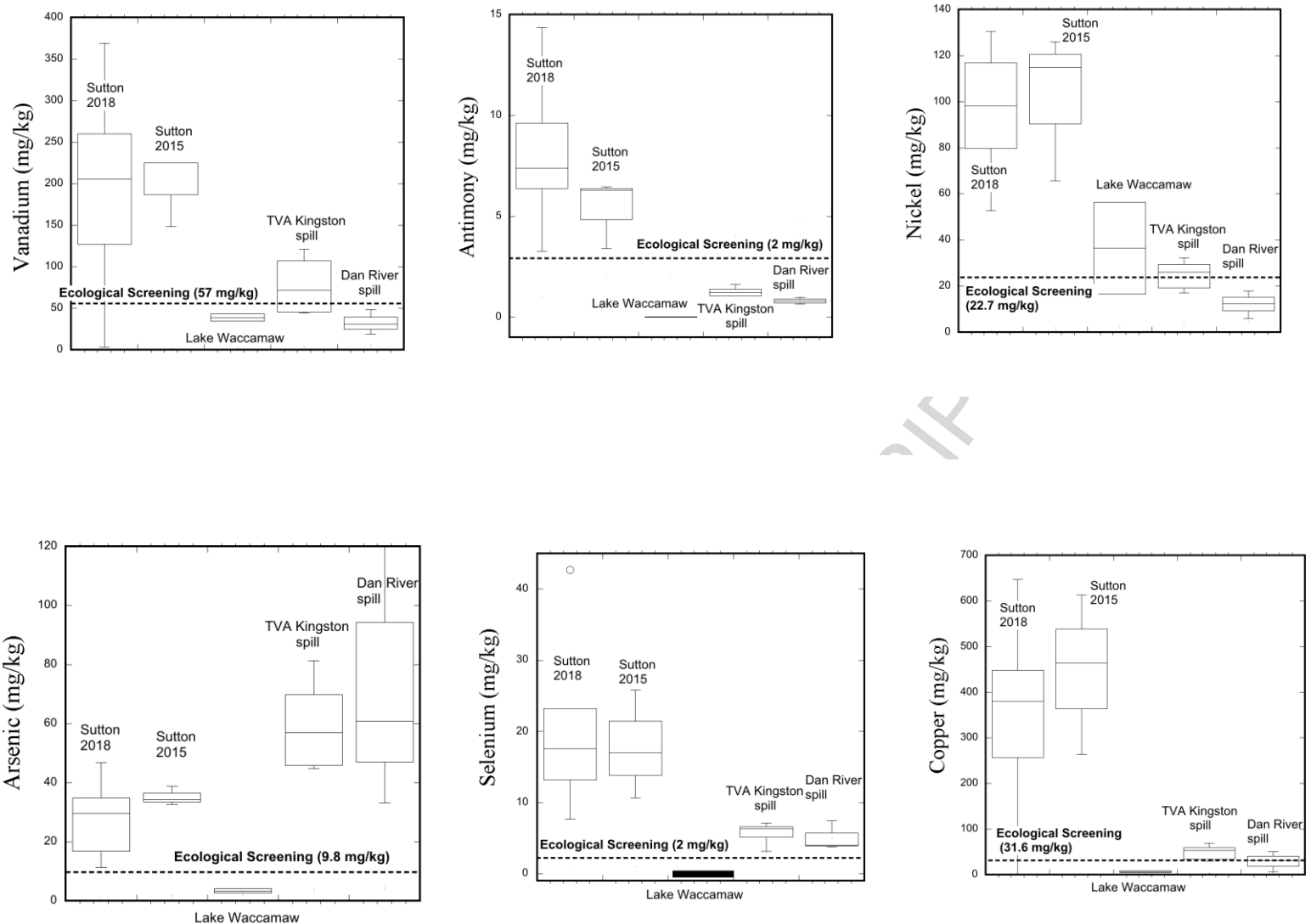


Figure 6: Box plots of selected metals with ecological impact measured in sediments collected from Sutton Lake in 2018 and 2015, Lake Waccamaw (reference lake), and in sediments impacted by the TVA Kingston (Tetra Tech Report, 2008) and Dan River (N.C. Water Quality Report, 2018) coal ash spills, compared to freshwater sediment screening benchmarks values for sediment toxicity used by U.S. EPA (MacDonald et al. 2000; MacDonald, 2003). Results from Sutton Lake sediments show high levels of metals comparable and in most cases higher than metals concentrations found in sediments impacted by the TVA Kingston and Dan River coal ash spills. The metals concentrations exceeded the acceptable screening thresholds for aquatic freshwater sediment toxicity used by U.S. EPA to define potential ecological impact (Long et al., 2000; 2006; MacDonald et al., 2000; MacDonald, 2003).

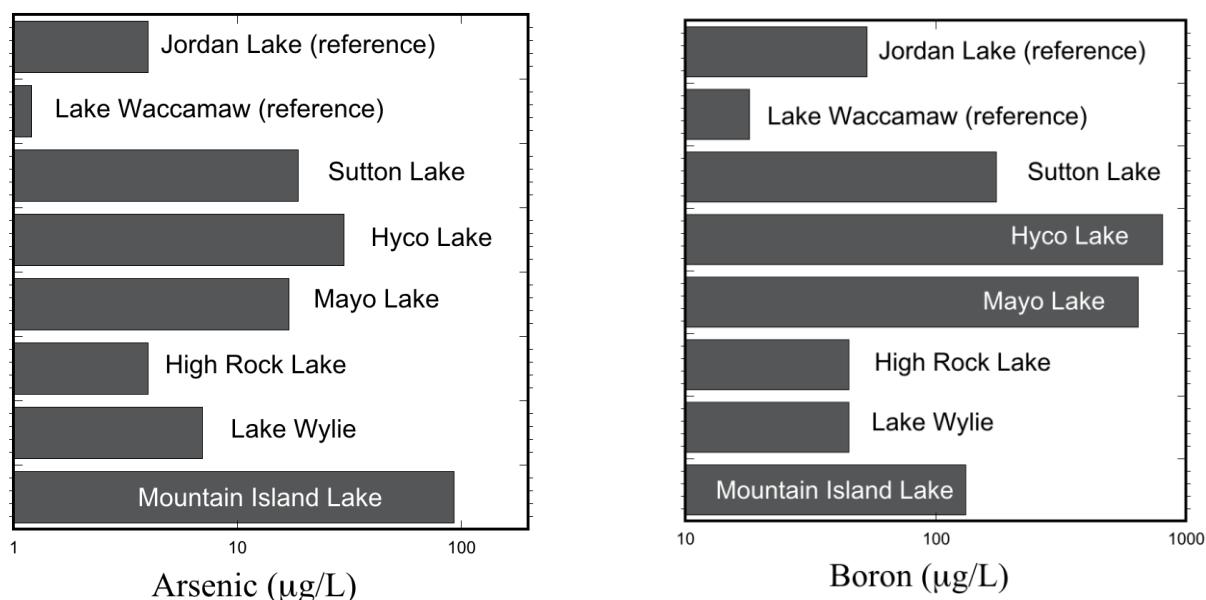


Figure 7: Concentrations of arsenic and boron in pore water from lakes located near CCR storage sites (Sutton, Hyco, Mayo, High Rock, Wylie, and Mountain Island (Brandt, 2018; Ruhl et al., 2012) compared to reference lakes without potential impact from nearby CCR storage sites (Waccamaw and Jordan). The location of the lakes is shown in Figure 1. The enrichment of As and B in pore water from bottom sediments in lakes located nearby CCR storage sites is suggested to be derived from mobilization of As and B from CCR solids that were spilled and accumulated in the lakes, as evidenced from the Sutton Lake case.

Sample Name	Li	Be	B	V	Cr	Mn	Co	Ni	Cu	As	Se	Rb	Sr	Mo	Ag	Cd	Sb	Ba	Tl	Pb	Th	U
	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg
Certified Value 1633c		16		286.2	258	240	42.9	132	173.7	186.2	13.9	117.42	901			0.758	8.56	1126		95.2	23	9.25
1633c	176.8	14.8	76.6	236.4	184.7	222.1	40.1	138.7	175.0	186.2	19.3	113.3	906.9	26.0	0.7	0.7	8.6	1098.3	6.6	101.1	22.1	9.0
1633c	170.5	14.7	75.7	229.4	177.4	216.7	38.9	129.0	167.8	181.0	18.8	112.4	894.0	25.4	0.6	0.6	8.4	1124.4	6.4	98.4	21.8	8.9
1633c	172.0	14.6	74.9	229.5	178.7	211.4	38.8	124.2	165.0	182.8	16.7	110.1	879.4	25.6	0.6	0.6	8.1	1069.3	6.5	98.3	21.4	8.9
1633c	174.1	14.4	76.4	228.3	180.3	216.3	38.4	124.0	165.9	182.0	15.7	111.6	875.3	25.5	0.6	0.7	8.2	1091.1	6.5	98.0	21.4	8.7
Mean	173.4	14.6	75.9	230.9	180.3	216.6	39.1	129.0	168.4	183.0	17.6	111.8	888.9	25.6	0.7	0.7	8.3	1095.8	6.5	99.0	21.7	8.9
Recovery%		91.4		80.7	69.9	90.3	91.1	97.7	97.0	98.3	126.9	95.2	98.7			88.6	97.3	97.3		104.0	94.3	95.8

Table 1: Concentrations of trace elements measured in fly ash standards (NIST 1633c) as compared to certified values. The recovery of the extraction procedure is the ratio between measured (n=4) and certified values, expressed in percent.

Sample	X_L (m ³ /kg)	Coal Ash (%)
Upstream		
CapeFear2	-5.85293E-09	0
SuttonLake2018		
Sutton1	1.38496E-07	5.0
Sutton5	1.5704E-06	38.7
Sutton7	1.30897E-06	27.7
Sutton6	6.06219E-09	0
Sutton3	6.21968E-06	89.0
Sutton4	6.8189E-07	21.0
CapeFearRiver2018 Downstream		
CapeFear31	5.58152E-09	0
CapeFear33	-3.74826E-09	0
SuttonLake2015		
Sutton1012015	1.03396E-06	28.6
Sutton1022015	4.49442E-06	58.6
Sutton1032015	3.28023E-07	16
ReferenceLake(2015)		
Waccamaw2	5.35389E-08	0
Waccamaw32	5.85811E-08	0

Table 2: Frequency dependent mass-specific magnetic susceptibility and percent coal ash counting from physical observation in sediments from different sources investigated in this study.

Sample	Li	Be	B	V	Cr	Mn	Co	Ni	Cu	Zn	As	Se	Rb	Sr	Mo	Ag	Cd	Sb	Ba	Tl	Pb	Th	U
Upstream																							
CapeFear2	2.2	0.2	0.6	1.9	1.6	28.5	0.9	0.9	0.8	19.5	0.1	BDL	19.2	31.1	0.0	0.0	0.0	BDL	191.6	0.1	7.3	1.2	0.3
SuttonLake2018																							
Sutton1	36.3	2.6	16.7	133.2	31.1	895.3	21.3	79.8	420.3	82.5	33.7	23.2	26.0	189.7	12.3	0.1	1.0	6.8	526.0	2.0	25.4	3.8	2.8
Sutton5	125.4	6.5	12.2	369.1	93.7	821.7	46.1	130.6	648.0	185.8	46.8	42.7	62.2	228.0	23.6	0.5	2.3	14.4	675.2	5.2	58.2	11.0	6.4
Sutton7	58.1	2.4	7.3	121.7	38.7	157.8	17.1	52.8	228.6	65.1	11.4	7.7	32.8	94.3	8.4	0.2	0.3	3.3	250.4	1.5	21.7	6.3	2.3
Sutton6	2.8	0.5	BDL	3.4	2.7	35.6	0.1	BDL	0.5	15.2	BDL	BDL	4.1	20.1	BDL	0.0	BDL	BDL	70.0	BDL	4.0	1.0	0.2
Sutton2	126.1	2.8	17.6	278.5	73.9	739.5	33.0	116.9	475.3	154.7	34.8	19.0	55.2	127.1	25.1	0.3	0.9	9.6	482.1	3.1	39.5	7.1	3.3
Sutton3	148.6	9.7	27.5	205.5	105.4	863.6	37.1	92.0	284.3	111.4	16.8	13.2	106.1	438.9	3.4	0.4	0.7	6.4	1000.1	2.3	39.4	17.7	6.0
Sutton4	137.3	6.3	22.5	242.0	89.6	801.6	35.1	104.4	379.8	133.1	25.8	16.1	80.7	283.0	14.3	0.4	0.8	8.0	741.1	2.7	39.5	12.4	4.7
CapeFearRiver-Downstream																							
CapeFear31	2.4	0.0	0.6	2.4	2.2	24.6	0.3	0.4	1.1	6.4	0.1	BDL	7.7	17.5	BDL	BDL	BDL	BDL	92.2	BDL	3.4	0.8	0.3
CapeFear33	3.1	BDL	14.1	5.1	2.4	19.2	1.2	10.2	6.4	26.6	1.4	BDL	10.8	25.9	BDL	0.1	BDL	BDL	92.3	0.1	5.1	0.3	0.2
SuttonLake2015																							
Sutton1012015	140.3	4.9	18.6	224.9	75.7	376.5	33.5	126.1	613.3	406.7	38.8	25.8	56.7	195.4	14.1	0.4	0.8	6.5	458.5	2.5	47.2	10.6	5.3
Sutton1022015	105.2	5.4	19.3	148.2	63.9	423.6	24.6	65.7	263.5	68.9	34.4	10.7	69.2	267.0	7.5	0.3	0.4	3.4	515.0	1.7	34.2	12.0	4.3
Sutton1032015	103.7	2.5	7.8	224.9	56.4	310.4	27.0	115.0	463.8	116.6	32.6	17.0	46.0	136.9	20.6	0.3	0.8	6.3	342.7	3.3	39.6	7.3	3.3
ReferenceLake(2015)																							
Waccamaw2	25.4	0.5	21.3	43.6	35.3	157.9	9.1	56.4	8.3	306.5	4.2	0.1	15.9	43.0	0.1	0.2	0.2	BDL	160.7	0.2	34.3	7.3	2.0
Waccamaw3	20.0	0.4	15.3	34.7	28.5	149.5	7.2	16.6	4.8	281.7	2.6	BDL	11.3	31.3	BDL	0.1	0.1	BDL	110.8	0.2	28.5	5.8	1.4

Table 3: Concentrations of trace elements in sediments from different sources investigated in this study.

Mean Values	n	Li	Be	B	V	Cr	Mn	Co	Ni	Cu	Zn	As	Se	Rb	Sr	Mo	Ag	Cd	Sb	Ba	Tl	Pb
Average Sutton 2018	7	90.7	4.4	17.3	193.3	62.2	616.4	27.1	96.1	348.1	106.8	28.2	20.3	52.4	197.3	14.5	0.3	1.0	8.1	535.0	2.8	32.5
Average Sutton 2015	3	116.4	4.3	15.2	199.4	65.3	370.2	28.4	102.3	446.9	197.4	35.3	17.8	57.3	199.8	14.1	0.3	0.7	5.4	438.7	2.5	40.4
Average Waccamaw 2015	2	22.7	0.5	18.3	39.1	31.9	153.7	8.1	36.5	6.6	294.1	3.4	0.1	13.6	37.2	0.1	0.2	0.2	0.0	135.8	0.2	31.4
NIST 633c (fly ash standard)	4	173.4	14.6	75.9	230.9	180.3	216.6	39.1	129.0	168.4	236.8	183.0	17.6	111.8	888.9	25.6	0.7	0.7	8.3	1095.8	6.5	99.0
Ratio Sutton Lake 2018/2015		0.78	1.02	1.14	0.97	0.95	1.67	0.96	0.94	0.78	0.54	0.80	1.14	0.91	0.99	1.03	0.84	1.51	1.50	1.22	1.14	0.81
Ratio Sutton Lake 2015/Lake Waccamaw		3.99	9.42	0.95	4.94	1.95	4.01	3.33	2.63	53.05	0.36	8.27	265.23	3.85	5.30	109.96	1.78	5.57	363.82	3.94	12.43	1.03
Ratio Sutton Lake 2018/Lake Waccamaw		5.12	9.20	0.83	5.09	2.05	2.41	3.48	2.80	68.10	0.67	10.34	232.65	4.21	5.37	106.58	2.12	3.69	242.85	3.23	10.94	1.28

Table 4: Mean values of trace-element concentrations measured in sediments from Sutton Lake collected in 2018 and 2015, Waccamaw Lake, Cape Fear River (upstream and downstream from Sutton Lake), and fly ash standard (NIST 1633c). The lower panel shows the ratios of different elements in sediments from different sources.

Variable	Value	Units
Lake Sutton Area	4,597,952.0	m ²
Sediment Depth	25.0	cm
Sediment Depth	0.3	m
Volume of Sediment	1,149,488.0	m ³
Volume of Sediment	1.15E+12	cm ³
Mean Solid Flow Rate	4.5	mg/L
Effluent Discharge Rate	4.6	mgd
Effluent Discharge Rate	1.74E+07	L per day
Effluent Discharge Rate	6.36E+09	L per year
TSS of Input Water	4.5	mg/L
Percentage of Coal Ash in Sediment from App State or chemistry data	10%	
Mass Input to Lake	2.86E+10	mg/yr
Mass Input to Lake	2.86E+07	g/yr
Density of Lake Sediment	2650	kg/m ³
Density of Lake Sediment	2.65	g/cm ³
Density of Coal Ash	1.6	g/cm ³
Amount of Sediment	2.93E+12	g
Time to fill	1.02E+05	years

Table 5: Mass-balance calculations for possible discharge of CCRs from routine discharge of CCR effluents from the Sutton Lake outfall. The mass- calculations were based on the discharge rates reported from Sutton lake outfall multiple by the total solids in the effluents (TSS) for obtaining the annual mass of CCRs discharged from regulated outfall in 2010 to 2013. That annual mass flux was compared to the mass of the upper 30 cm of Sutton Lake bottom sediments. Given the known flux rate and the lake sediments volume (converted to mass) the time to fill the lake sediments with CCR was calculated. Data for effluents discharge and concentrations of total suspended solids (TSS) are from mean values calculated from NC DEQ reported data (2010-2013).

Sample		Li	Be	B	V	Cr	Mn	Co	Ni	Cu	Zn	As	Se	Rb	Sr	Mo	Ag	Cd	Sb	Ba	Tl	Pb	Th	U	87Sr/86Sr ratio
Upstream	water leaching																								
Cape Fear#2		27.4	0.1	175.8	29.3	35.3	235.1	31.0	115.2	9.6	420.4	3.6	2.5	7.5	58.0	1.9	BLD	0.1	0.7	70.8	0.1	21.3	0.0	0.7	
Sutton Lake#2018																									
Sutton#1		233.1	BLD	1646.8	455.9	43.9	14669.6	130.5	344.7	156.9	549.0	317.6	76.9	121.2	4350.6	342.9	0.1	1.9	126.2	1010.8	32.6	34.6	0.0	0.8	0.709931
Sutton#5		104.7	BLD	2975.8	316.8	39.6	6678.9	30.0	224.5	237.1	440.6	157.1	129.9	159.2	2563.0	697.8	BLD	2.6	86.3	1171.5	46.8	10.9	0.0	1.1	0.710375
Sutton#7		70.2	0.9	842.7	265.5	32.5	2643.3	26.9	168.6	156.8	444.7	77.4	49.7	112.3	1969.3	1008.8	BLD	1.7	86.5	692.6	26.8	9.6	0.2	1.2	
Sutton#6		61.5	0.5	514.9	77.8	70.1	409.4	15.9	314.8	56.9	1352.8	21.2	0.0	6.0	121.2	9.1	BLD	0.0	1.4	62.1	0.8	10.0	0.3	1.3	
Sutton#2		108.4	0.3	1279.8	1947.6	28.6	3834.2	32.3	245.1	196.5	418.5	240.2	131.0	124.8	2458.0	4619.5	BLD	6.1	408.8	868.8	26.2	7.0	0.0	1.0	0.710025
Sutton#3		94.6	BLD	870.2	173.3	75.2	2848.6	32.1	405.6	368.6	1461.7	38.9	56.9	128.9	1811.2	158.3	0.1	0.5	72.9	612.2	19.0	11.4	0.2	0.5	0.709997
Sutton#8		120.0	BLD	977.2	553.3	24.3	4758.8	30.7	185.2	178.9	402.2	253.6	131.0	134.6	1787.1	2396.3	0.0	3.5	133.4	889.5	39.8	3.6	0.0	0.4	
Cape Fear River#downstream																									
Cape Fear#1		74.6	BLD	645.1	60.1	114.3	397.4	38.0	383.5	32.6	1457.5	12.8	2.1	7.3	188.6	7.8	0.1	0.0	2.6	104.5	0.1	33.2	1.4	1.0	
Cape Fear#3		737.1	0.3	527.1	12.3	67.6	131.9	15.9	317.1	12.4	1402.6	1.6	0.0	7.6	49.0	0.0	BLD	0.0	0.6	35.3	0.2	4.6	0.0	0.4	
Sutton Lake#2015																									
Sutton#1#2015		1280.0	3.3	1652.9	135.5	44.8	71055.2	546.9	1253.8	1805.2	1243.3	73.9	150.5	271.2	14269.1	32.5	BLD	27.9	38.6	1179.3	105.1	49.7	0.0	1.6	0.710009
Sutton#2#2015		1205.7	0.0	1856.0	18.7	91.3	95342.0	399.4	1294.2	648.4	2050.3	49.6	69.3	166.2	11664.5	11.7	BLD	19.7	17.4	1172.1	59.2	66.3	0.0	0.0	0.710171
Sutton#3#2015		3312.8	11.0	1580.3	284.3	96.7	100334.5	3238.0	8618.5	2208.1	8478.4	88.5	137.2	311.8	27409.9	71.4	BLD	97.0	68.5	1089.1	232.0	72.8	0.0	1.6	0.709932
Reference Lake																									
Waccamaw#2		17.0	BLD	430.8	15.4	23.5	7235.8	15.9	128.2	12.4	538.0	6.8	0.0	67.7	1940.8	1.4	BLD	1.3	3.3	655.2	6.9	35.4	0.0	0.4	0.709563
Waccamaw#3		57.7	1.3	938.4	122.6	120.5	8887.0	29.4	441.9	63.8	1687.1	22.1	5.7	168.9	2366.9	2.3	0.1	3.7	4.2	584.1	17.4	97.0	4.1	4.6	0.709527
Upstream	Acid leaching																								
Cape Fear#2		59.1	48.8	142.0	279.5	193.6	4773.2	412.4	360.3	103.7	3448.9	18.2	10.6	44.2	321.2	2.0	0.5	9.6	2.7	2185.8	4.9	431.9	26.2	24.0	
Sutton Lake#2018																									
Sutton#1		9966.7	1407.2	3322.1	116927.9	6812.4	411625.7	14235.4	56084.4	293384.2	40687.9	24992.8	4733.3	710.9	103609.4	2552.8	6.4	770.7	585.8	75727.9	1711.1	19614.4	378.6	2342.9	
Sutton#5		46990.2	2477.5	4436.6	331088.3	29928.1	352040.5	27945.1	78901.8	495894.6	98264.8	34733.3	5125.7	1894.3	74491.4	8941.9	51.5	1641.9	1065.4	67471.7	4012.3	37029.8	2286.3	4079.6	
Sutton#7		31958.3	1547.7	1831.9	139422.8	14542.9	181449.8	19065.0	50668.2	264335.0	40663.2	13977.3	1977.9	1285.5	45311.6	5407.0	23.7	686.1	571.1	76665.9	2076.0	22182.2	1029.4	2015.6	
Sutton#6		308.5	12.9	550.2	483.5	144.2	4944.2	100.7	598.6	1186.3	2418.3	165.3	9.6	35.9	795.5	43.9	0.5	2.7	9.7	1640.7	10.6	389.6	9.3	25.3	
Sutton#2		23308.5	1061.9	1480.7	194479.3	12392.6	216724.1	14367.9	61485.4	288295.9	41669.5	20429.1	3024.0	1823.4	69854.3	7378.1	26.1	644.9	550.2	38386.7	2212.4	25227.6	1452.2	1949.2	
Sutton#3		27469.9	1441.5	2473.0	93395.0	6924.7	331597.4	12194.4	29166.2	159662.5	38228.7	6600.3	1444.1	1505.8	83212.8	794.0	7.6	384.8	486.4	124617.0	1073.6	16772.5	669.3	1631.8	
Sutton#8																									
Cape Fear River#downstream																									
Cape Fear#1		289.2	6.8	187.6	236.5	138.0	3970.0	66.7	261.0	268.7	1468.2	75.7	9.7	59.8	885.7	16.9	0.5	2.2	7.3	1561.3	4.2	288.1	30.7	17.9	
Cape Fear#3		108.6	29.2	463.8	207.5	229.2	4334.8	415.7	655.1	124.7	4610.3	28.1	30.7	86.5	464.8	3.4	1.2	14.9	2.6	2684.6	4.1	366.3	32.4	28.1	
Sutton Lake#2015																									
Sutton#1#2015		29674.0	1183.2	3910.2	99691.2	7547.1	383623.7	13680.7	30938.5	193075.5	39174.5	24087.1	2314.7	1211.2	175682.9	2332.8	3.3	384.4	475.5	119693.7	997.2	14267.0	553.7	1505.0	
Sutton#2#2015		29674.0	1183.2	3910.2	99691.2	7547.1	383623.7	13680.7	30938.5	193075.5	39174.5	24087.1	2314.7	1211.2	175682.9	2332.8	3.3	384.4	475.5	119693.7	997.2	14267.0	553.7	1505.0	
Sutton#3#2015		3312.7	494.5	1133.6	11272.5	6861.6	100811.6	4416.6	5894.1	3666.6	36950.9	743.4	811.5	681.4	18597.1	65.1	4.4	418.4	25.4	49769.3	159.2	27729.3	337.5	827.9	
Reference Lake																									
Waccamaw#2		3686.5	584.8	1164.8	11085.4	6586.3	105005.7	4832.2	5549.7	3272.5	38368.3	592.2	772.6	764.9	17425.0	51.0	2.5	333.2	24.8	45677.6	129.7	26490.9	254.6	728.6	
Waccamaw#3		3312.7	494.5	1133.6	11272.5	6861.6	100811.6	4416.6	5894.1	3666.6	36950.9	743.4	811.5	681.4	18597.1	65.1	4.4	418.4	25.4	49769.3	159.2	27729.3	337.5	827.9	

Table 6: Concentrations of trace elements measured in water and acid (1N nitric acid) leachates extracted from sediments in Sutton Lake collected in 2018 and 2015, Waccamaw Lake, and Cape Fear River (upstream and downstream from Sutton Lake).

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Graphical abstract

